Two-Photon Emission from Semiconductors

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We report, to the best of our knowledge, the first experimental observation of spontaneous two-photon emission from semiconductors. The overall two-photon emission power is only 4 orders of magnitude smaller than the fundamental one-photon emission power due to the continuous energy band structure of semiconductors. The measured wide-band two-photon emission spectrum is surprisingly blue-shifted in contrast to the two-photon emission from discrete-level atomic systems. This shift can be accounted for by the second-order matrix element k-dependence in semiconductors and the measured spectrum shape appears to be in good agreement with our calculations.

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In the process of two-photon emission the transition between quantum levels occurs via simultaneous emission of two photons. The calculation of the probability for such a transition requires summing over all bound and continuum states of the system [1]. The individual photon energies from these decays form a continuum, while their sum is equal to the transition energy. This phenomenon is of a great importance for applications in astrophysics, contributing to the continuum radiation from planetary nebulas [2], and atomic physics due to the emitted spectrum dependence on the entire system quantum level structure [3]. Furthermore, the earliest attempts to produce polarization-entangled photons were performed via 2-γ photon decay in positronium [4], highlighting most vividly the non-locality of quantum mechanics through violation of Bell's inequalities [5]. Recently semiconductor two-photon emission was proposed as a compact room-temperature high-rate source of polarization-entangled photons [6]. Moreover efficient electrically-driven photon-number sources can be easily implemented employing semiconductor two-photon process as heralded photon emission similar to the non-degenerate parametric down conversion [7] but with much higher efficiency and simpler design requirements.

Two-photon transition is described by a second order process in the time-depended perturbation theory, and thus it is much weaker than the first-order one-photon process. Therefore observations of multi-photon spontaneous decays have so far been restricted to a few atomic transition cases, where the lowest-order transition is forbidden by selection rules [8] or suppressed by a cavity [9]. First-order one-photon atomic transitions between discrete energy levels have a nearly discrete spectrum, while their two-photon spectrum is continuous and centered at about half the one-photon transition frequency [10].

Semiconductors can have very high carrier densities, making even the weak second-order transitions measurable, and their energy level structure consists of continuous energy bands, causing the one-photon emission spectrum to have finite band-width. Moreover, the whole complex energy-band structure is probed through the transition intermediate states, thus strongly affecting the second order processes

spectrum. There has been a substantial theoretical and experimental effort to investigate the multi-photon absorption effects in semiconductors [11, 12], however, two-photon emission from semiconductors, to the best of our knowledge, has been neither observed nor theoretically analyzed yet.

Here we report the first experimental observation of spontaneous two-photon emission from semiconductors with the wide-spectrum two-photon emission power being only 4 orders of magnitude smaller than the fundamental one-photon emission and surprisingly blue-shifted in contrast to the two-photon emission from discrete-level atomic systems. This shift can be accounted for by the electron crystal momentum dependence (k-dependence) of the second-order matrix elements in semiconductors.

Two-photon emission rate from a semiconductor is calculated by a second order process in the time-dependent perturbation theory, similar to that of the two-photon transition in a discrete-level atom, and the emission rate at a specific wavelength in atomic units is [1, 10].

$$\frac{dW}{d\omega_1} = \frac{\omega_1 \omega_2}{(2\pi)^3 c^2} |M|^2 \, d\Omega_1 d\Omega_2 \tag{1}$$

where ω_i is the photon energy, $d\Omega_i$ is the solid angle for the i-th photon emission and M is the second-order matrix element given by:

$$M = \sum_n \left(\frac{\langle f | \hat{p}\varepsilon_2 e^{-ik_2 x} | n \rangle \langle n | \hat{p}\varepsilon_1 e^{-ik_1 x} | i \rangle}{E_i - E_n - \hbar\omega_1} + \frac{\langle f | \hat{p}\varepsilon_2 e^{-ik_2 x} | n \rangle \langle n | \hat{p}\varepsilon_1 e^{-ik_1 x} | i \rangle}{E_i - E_n - \hbar\omega_2} \right) \tag{2}$$

where p is the momentum operator, ε is the photon polarization, i, n and f are the initial, intermediate and final electron states respectively. A number of experiments and calculations for two-photon emission from various discrete-level systems have

demonstrated a wide-band continuous spectrum having a maximum at twice the resonant wavelength [1, 10].

The fundamental difference between two-photon processes in semiconductors and in discrete-level atomic systems lies in the solid-state unbounded electron wavefunctions with definite crystal momentum in contrast to the bounded atomic states. Continuum of unbounded electron states in every band of the semiconductor introduces a great number of available intermediate states, making semiconductor two-photon process much more efficient versus the atomic discrete-level systems. Furthermore, the definite crystal-momentum states allow using the intra-band transitions as part of the two-photon second-order calculations causing the matrix element to be k-dependent unlike for the inter-band transitions.

We calculate the shape of the two-photon emission spectrum from GaInP quantum wells (QWs) at room temperature with the injected current of 200mA using only the valence and the conduction bands and neglecting other bands due to their energy remoteness. The second-order process probability is calculated using the above bands as the intermediate state combining an intra-band and an inter-band transition matrix elements, following the models used for calculating two-photon absorption in semiconductors [12, 13]:

$$\begin{aligned} \left\langle f \left| \hat{p}\varepsilon_2 e^{-ik_2 x} \right| n \right\rangle \left\langle n \left| \hat{p}\varepsilon_1 e^{-ik_1 x} \right| i \right\rangle_1 &= \frac{m}{m_c} \hbar k \sqrt{\frac{1}{2}} p \\ \left\langle f \left| \hat{p}\varepsilon_2 e^{-ik_2 x} \right| n \right\rangle \left\langle n \left| \hat{p}\varepsilon_1 e^{-ik_1 x} \right| i \right\rangle_2 &= \frac{m}{m_v} \hbar k \sqrt{\frac{1}{2}} p \end{aligned} \tag{3}$$

where p is the k-independent inter-band part of second order matrix element:

$$p^2 = \frac{3m^2}{2} \left(\frac{1}{m_c} - \frac{1}{m} \right) \frac{E_g \left(E_g + \Delta \right)}{3E_g + 2\Delta} \tag{4}$$

E_g is the energy gap, k is the electron crystal momentum , Δ is the spin-orbit splitting of the valence band, m is the free-space electron mass and m_v and m_c are the valence band and the conduction band effective masses respectively. The calculations show a significant shift of the emission maximum towards shorter wavelengths in the semiconductor due to the matrix element k-dependence, whereas in a discrete level system with the energy separation equal to the average carrier energy in the semiconductor the emission maximum is located at twice the resonant wavelength as expected (Fig. 1).

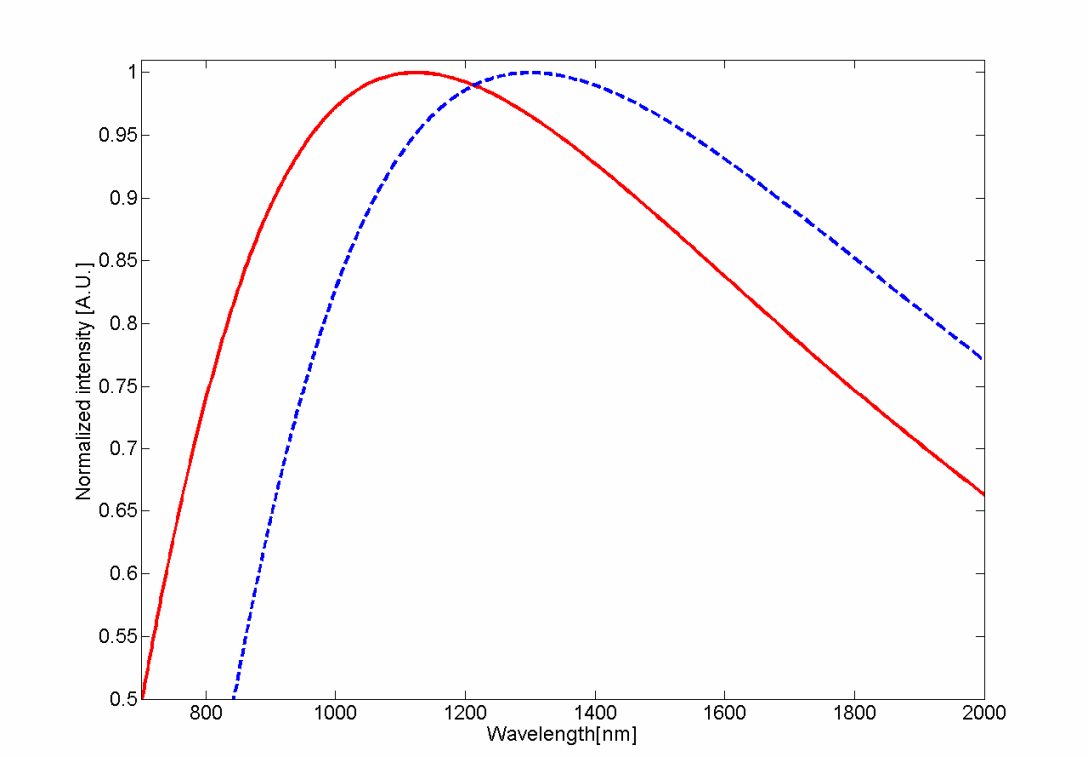

Figure 1. Calculated two-photon emission spectrum from 670nm-wavelength GaInP QWs – solid line. The dashed line is the two-photon emission spectrum from a discrete level system with the energy separation equal to the average carrier energy in GaInP QWs.

In the experiment, semiconductor samples were used consisting of 4 periods of compressively strained 50Å $Ga_{0.45}In_{0.55}P$ QWs with one-photon emission spectrum centered at 670nm separated by 55Å $(Al_{0.5}Ga_{0.5})_{0.51}In_{0.49}P$ barriers, ~1.1μm AlGaInP cladding and a 300nm GaAs cap layer having near 890nm one-photon emission

wavelength. The short-wavelength (670nm) high-power GaInP QW structure enables the detection of the double-wavelength two-photon emission maximum by standard InGaAs photo-receivers. Using longer-wavelength infrared (IR) emitting materials, used in optical communications, would result in two-photon emission having a maximum at a wavelength undetectable by InGaAs, whereas shorter-wavelength GaN devices cannot provide high enough output powers. The lateral light confinement was achieved by a 4μm-wide ridge waveguide, realized by standard processing in order to enhance the emitted photons collection efficiency. The structure was electrically pumped below the lasing threshold, which was increased to above 200mA by applying an anti-reflection coating on the facets, in order to enable higher carrier population inversion. Spatial distribution of the wide-band IR two-photon emission intensity was centered near the QWs, whereas the 890nm narrow-band emission was located in the GaAs cap layer region (Fig.2).

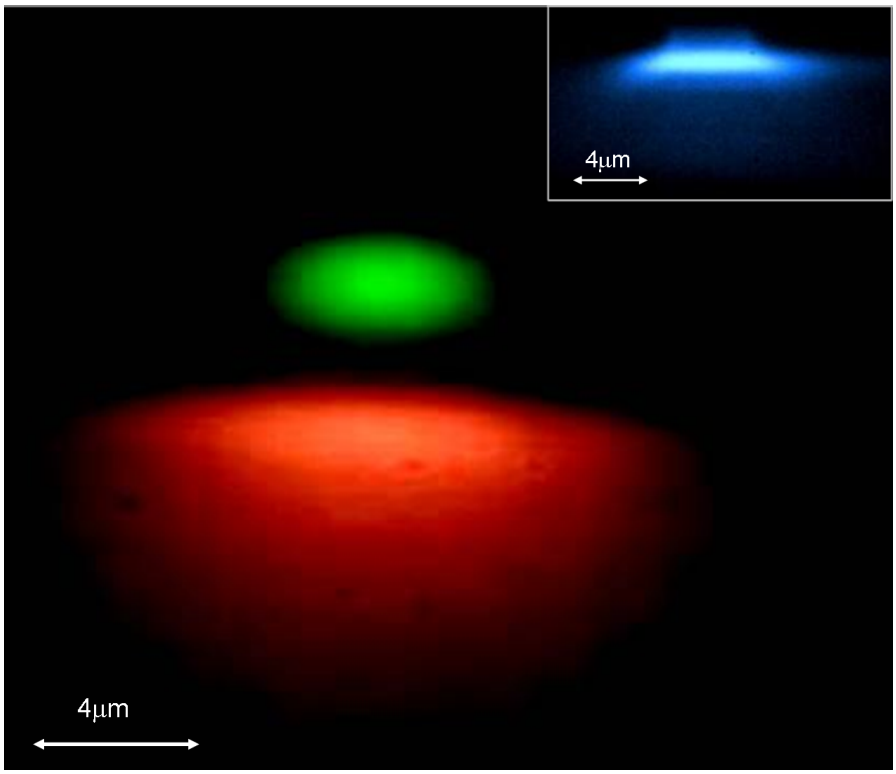

Figure 2. Experimental false color IR emission imaging of the facet of the 670nm-wavelength GaInP QWs based WG. The upper lobe (green) is the 890nm emission from the GaAs cap layer and the lower lobe (red) is the wide-band two-photon emission from GaInP QWs at ~200mA injection current. The inset is a snapshot of the visible 670nm emission from the same structure at ~10mA injection current.

IR spectrum measurements were conducted using an Acton-Research monochromator equipped with a New-Focus IR femtowatt photo-receiver and a lock-in amplifier. The spectrum for the two-photon emission was very wide-band as expected, nevertheless its maximum appears at a wavelength shorter than twice the single-photon wavelength (Fig. 3). This result in good agreement – qualitatively, with the calculations (Fig. 1). The overall two-photon emission intensity measured is about 4 orders of magnitude lower than the fundamental one-photon emission.

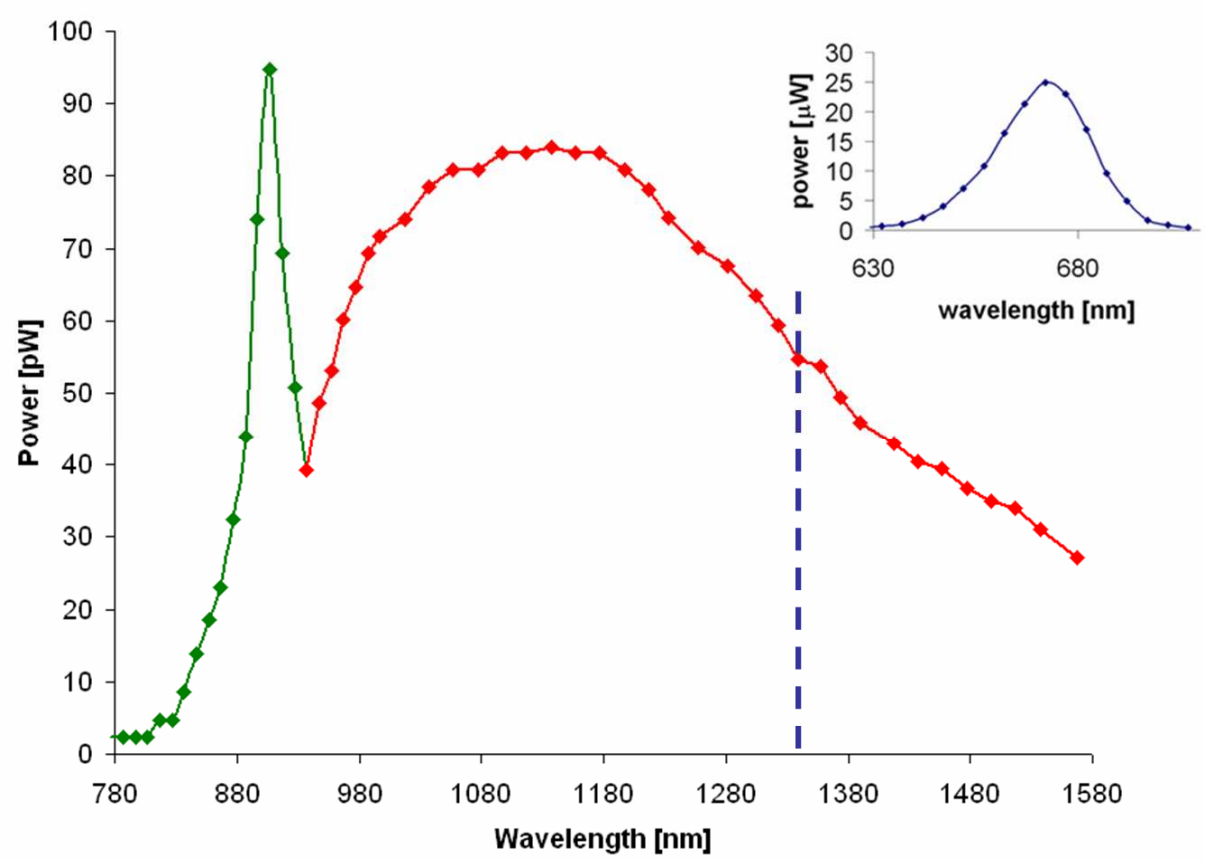

Figure 3. Measured IR emission spectrum from 670nm-wavelength GaInP QWs. The narrow-band 890nm peak (green) is the one-photon emission from the GaAs cap layer and the wide-band two-photon emission (red) is from the GaInP QWs. The dashed line indicates twice the one-photon QW emission wavelength ~1340nm. The inset is the spectrum of the one-photon visible 670nm emission.

In conclusion we have demonstrated an experimental observation of spontaneous two-photon emission from a QWs semiconductor structure, which as far as we know, has been neither observed nor theoretically analyzed before. The measured wide-band two-photon emission spectrum is blue-shifted in contrast to the two-photon emission from discrete-level atomic systems which may be attributed to the k-dependence of the second-order matrix element in semiconductors and the measured spectrum shape is in good agreement with our calculations. The overall two-photon emission power is only 4 orders of magnitude smaller than the fundamental one-photon emission due to the continuous energy band structure of semiconductors. Such high two-photon emission intensity may help exploiting this fundamental physical phenomenon for practical uses in both science and engineering.

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